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Characterization of the performances of an innovative heat-exchanger/reactor

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ABSTRACT

The use of heat exchanger/reactors (HEX/reactors) is a promising way to overcome the barrier of poor heat transfer in batch reactors. However to reach residence time long enough to complete the chemistry, low Reynolds number has to be combined with both a plug flow behaviour and the intensification of heat and mass transfers. This work concerns the experimental approach used to characterize an innovative HEX/reactor. The pilot is made of three process plates sandwiched between five utility plates. The process stream flows in a 2 mm corrugated channel. Pressure drop and residence time distribution characterizations aim at studying the flow hydrodynamics. Identified Darcy correlations point out the transition between laminar and turbulent flow around a Reynolds number equal to 200. Moreover the flow behaves like a quasi-plug flow ($Pe > 185$). The heat transfer and mixing time have also been investigated. The ratio between the reaction kinetics and the mixing time is over 100 and the intensification factor ranges from 5000 to 8000 $kW\,m^{-3}\,K^{-1}$. As a consequence, no limitations were identified which allows the implementation of an exothermic reaction. It has been successfully performed under severe temperature and concentration conditions, batchwise unreachable. Thus, it highlights the interest of using this continuous HEX/reactor.

1. Introduction

Among the different ways of process intensification [1], the batch-to-continuous transposition is one of the most classical, provided that the reactor has to behave as plug flow reactor. The main issues in such apparatuses are to intensify heat and mass transfers while operating at low-Reynolds flows usually characteristic of laminar flows. Indeed low fluid velocities, i.e. high residence times, are required to complete the chemistry although high heat and mass transfer coefficients are usually expected in turbulent flows. One solution to remove this barrier is to work with heat-exchanger/reactors [2–4]. These apparatuses combine a heat-exchanger and a reactor in the same unit. This allows an accurate control of the operating temperature which is one of the main barriers concerning the implementation of exo- or endo-thermal reactions in classical batch reactors. Moreover the corrugation of the reaction channel leads to the heat and mass transfers intensification while maintaining low Reynolds numbers [5–9]. Thus, by using heat exchanger/reactors many benefits are expected such as waste reduction, energy and raw materials saving, yield and selectivity increase, and cost reduction.

The aim of this paper is to investigate the performances of the continuous heat exchanger/reactor developed in the frame of the RAPIC R&D project supported by the French Agency ANR [7,10]. The originality of this project which started in December 2007 and lasted 42 months was to develop an innovative low-cost heat-exchanger/reactor (HEX reactor) for the 10 $kg\,h^{-1}$ nominal flowrate. The development strategy to reach this goal was to take into consideration the implementation constraints of an industrial exothermal reaction while being as close as possible to mature technologies of heat-exchanger. A complementary consortium was thus setup comprising one end-user (Rhodia Chemicals), one heat-exchanger manufacturer (Fives Cryo), two French laboratories active in basic research on process engineering (LGC) and thermo-hydraulic engineering (LTN), and the Atomic and Alternative Energy Commission (CEA/LITEN) which handles the project coordination, the component design, and the reaction plates manufacture.

The first part of this paper deals with the presentation of the heat exchanger/reactor and the experimental set-up used to characterize it. Its hydrodynamic performances are then evaluated from pressure drop, residence time distribution and mixing times measurements. The object of the third part is the study of the reactor

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Nomenclature

A	heat exchange area (m^2)
C_p	heat capacity ($J\,kg^{-1}\,K^{-1}$)
d_{bends}	distance between two successive bends (m)
d_h	hydraulic diameter (m)
De	Dean number
D_{ax}	axial dispersion coefficient ($m^2\,s^{-1}$)
D_h	hydraulic diameter (m)
E_a	activation energy ($J\,mol^{-1}$)
$f_{corrugation}$	corrugation frequency (s^{-1})
F_p	process volume flowrate ($L\,h^{-1}$)
k_0	reaction rate constant ($L\,mol^{-1}\,s^{-1}$)
L	process channel length (m)
L_{total}	total process channel length (m)
m_{dw}	mass of water equivalent to the Dewar vessel (kg)
m_w^0	mass of water initially introduced in the Dewar vessel (kg)
m_p	mass of sample poured in the Dewar vessel (kg)
M_p	process mass flowrate ($kg\,h^{-1}$)
M_u	utility mass flowrate ($kg\,h^{-1}$)
N_i^0	initial molar flowrate of the limiting reactant ($mol\,s^{-1}$)
n_i^0	initial mole number of the limiting reactant (mol)
Pe	Peclet number
Q_{losses}	heat losses
Q_p	heat exchanged by the process side (W)
$Q_{r,total}$	total heat reaction (W)
R	gas constant ($J\,mol^{-1}\,K^{-1}$)
R_c	channel curvature radius (m)
Re	Reynolds number
Re_p	process Reynolds number
Re_u	utility Reynolds number
t	time (s)
t_m	mixing time (s)
t_r	residence time (s)
T_{dw}^0	temperature in the Dewar vessel just before introducing the reactive media (°C)
$T_{eq,dw}$	temperature at equilibrium in the Dewar vessel
$T_{p,in}$	process inlet temperature (°C)
$T_{p,out}$	process outlet temperature (°C)
$T_{u,in}$	utility inlet temperature (°C)
$T_{u,out}$	utility outlet temperature (°C)
u	process fluid velocity ($m\,s^{-1}$)
U	global heat transfer coefficient ($W\,m^{-2}\,K^{-1}$)
V	process channel volume (m^3)

Greek letters

ΔH_r	reaction heat ($J\,mol^{-1}$)
ΔP	pressure drop (Pa)
ΔT_{ml}	logarithmic mean temperature (°C)
Λ	Darcy coefficient
μ	viscosity (Pa s)
ρ	density ($kg\,m^{-3}$)
χ	conversion rate

thermal behaviour through the investigation of thermal profiles in the process channel during experiments consisting in cooling a hot process fluid by the utility side. Finally the heat removal ability of the heat exchanger/reactor is tested through the implementation of the highly exothermic reaction of sodium thiosulfate oxidation by hydrogen peroxide.

Table 1
Geometrical properties of the heat-exchanger/reactor.

	Process stream	Utility stream
Number of parallel channels	1	16
Number of plates for each stream	3	4
Individual channel width (mm)	2.0	2.0
Individual channel depth (mm)	2.0	2.0
Individual channel length, L_{total} (mm)	6700	
Hydraulic diameter, D_h (mm)	2.0	2.0
Total fluid volume (mm^3)	26.85	114.1
Metal thickness between streams (mm)	2	2

2. The heat-exchanger reactor

The process channel design has been optimized in the frame of the Rapic project in order to find a compromise between the reactor performances (heat transfer and mixing), the pumping power, the compactness and the manufacturing costs. Lab-scale mock-ups have been implemented to study the influence of the geometrical parameters (curvature radius, straight length between two bends, aspect ratio and bend angle) on the heat and mass transfer performances, the pressure drops and the residence time distribution. The whole approach is detailed in Ref. [9]. From both these works and the specifications of the End-User involved in the Rapic project, a corrugated geometry has been selected [7,10]. The pilot has been manufactured in accordance with the results of the geometry optimization. It consists in three reactive plates sandwiched between four utility plates. The reactive plates as well as the utility plates have been engraved by laser machining to obtain 2 mm square cross-section channels. Both reactive and utility channels designs are presented in Fig. 1(a) and (b), and their characteristics are detailed in Table 1.

The reactor material is 316L stainless steel and the different plates have been assembled by hot isostatic pressing (HIP) [7,10,11]. After assembly the reactor has a 32 cm height, a 14 cm width, a 3.26 cm thickness, and a mass of 10.84 kg, which makes it a very compact HEX reactor. The reactor has been dimensioned for a flowrate range from 1 to 15 $kg\,h^{-1}$, which corresponds to residence times ranging from 7 to 40 s.

3. The experimental setup

A schematic representation of the experimental set-up used to investigate the reactor performances is presented in Fig. 2. It involves two distinct fluid streams which enable to feed respectively the reactive channel or the utility circuit. The process line is equipped with two pumps and the utility circuit with a third one. Each pump is equipped with a mass flowmeter. Each process pump can be fed with its own reactant or by distilled water. The utility circuit is supplied by distilled water as coolant. The utility line temperature is fixed thanks to a thermally controlled bath located just upstream its entrance in the reactor.

The respective temperatures of the utility inlet and outlet are measured thanks to Pt100 sensors. For the reactive channel the inlet and outlet temperatures are measured thanks to K-type thermocouples. During experiments the different temperatures and flowrates are recorded.

Other specific sensors are added for the different steps of this study. They are presented and detailed in the related parts of this paper.

4. Methods, results and discussion

Before implementing a test reaction, the first part of these works aims at characterizing the hydrodynamic behaviour (pressure drop and residence time distribution) and the transfer mechanisms